BIODEGRADATION OF MANGANESE IN SEQUENCING BATCH REACTOR

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A thesis submitted in fulfillment
of the requirements for the award of the degree of
Bachelor of Chemical Engineering

Faculty of Chemical & Natural Resources Engineering
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MAY 2009

ABSTRACT

This thesis is about the biodegradation of manganese in sequencing batch reactor. Biodegradation of manganese in sequencing batch reactor (SBR) is a new invention treatment method that gives a lot of advantage in term of lowering operation and maintenance cost. The works done in this research explore the effects of different loading rate (LR) on the water quality parameter. For this research, biofilm process was selected as a treatment method which is develop in the SBR. At the beginning of the experiment, the mixed cultured was collected from drain and acclimatized in 10 liter reactor and monitoring the growth by using suspended solid (SS) test. The mixed cultured was acclimatized within two months by fed with 1mg/L manganese. For the treatment process, the mixed culture from acclimatization reactor will be transferred to the treatment reactor. The manganese treatment process will be carried out by controlling the loading rate. The hydraulic retention time (HRT) selected for this research is 5 days. The experiment will be run within two weeks. Within this period, the collection of the sample will be done for everyday as well as the addition of the simulated wastewater to the treatment reactor. The data shown that the lowest organic loading rate which is 3mg/L.d giving the highest percent of manganese removal. The highest manganese removal is 65% while the highest value of chemical oxygen demand (COD) removal is 72% at 3.5 mg/L.d. By using the Design Expert, the predicted value will represent the optimization results for this research. The value for optimization is at lowest LR which is 3 mg/L.d. By using lowest LR, the treatment will get 43% of COD removal and 57% manganese removal. The optimization of concentration of suspended solid is 381 mg/L.

ABSTRAK

Tesis ini adalah berkaitan dengan penguraian mangan secara biologi di dalam reaktor sesekumpul berjujukan. Penguraian secara biologi di dalam reaktor sesekumpul berjujukan adalah satu kaedah rawatan baru yang memberikan banyak kelebihan dari aspek pengurangan kos operasi dah kos penyelenggaraan. Kerja-kerja yang dijalankan di dalam kajian ini menyingkap perbezaan kesan kadar beban terhadap kualiti parameter air. Untuk kajian ini, kaedah rawatan yang dipilih adalah biofilem digabungkan dengan reaktor sesekumpul berjujukan. Pada permulaan eksperimen ini, kultur campuran diambil dari longkang dan dimasukkan dalam reaktor 10 liter untuk proses aklimitasi seterusnya pertumbuhan kultur campuran itu dipantau melalui ujian pepejal terampai. Proses penyesuaian kultur campuran tersebut di jalankan untuk tempoh masa dua bulan dengan memberi makan mangan kepada kultur campuran itu pada kepekatan 1 mg/L. Untuk proses rawatan, kultur campuran itu dipindahkan dari reaktor aklimitasi ke reaktor rawatan. Proses rawatan mangan dijalankan dengan mengawal nilai kadar beban. Masa penahanan hidraulik yang dipilih untuk kajian ini adalah 5 hari. Eksperimen ini dijalankan selama dua minggu. Dalam tempoh ini, sampel diambil dan tambahan sisa air buatan juga dilakuakan ke dalam reaktor yang mana proses ini dijalankan setiap hari. Data yang diperolehi menunjukkan bahawa kadar beban yang terendah iaitu 3 mg/L.h memberikan peratus penyingkiran mangan yang paling tinggi. Peratus penyingkiran paling tinggi adalah 65% manakala peratus penyingkiran tertinggi untuk permintaan oksigen kimia yang tertinggi adalah 72% pada 3.5 mg/L. Semua data yang diperolehi dimasukkan ke dalam perisian Design Expert untuk meramal nilai optimum bagi kajian ini. Kajian ini memperolehi nilai optimum pada kadar beban 3 mg/L.h. Dengan menggunakan kadar beban yang paling rendah, rawatan ini akan memperolehi penyingkiran permintaan oksigen kimia sebanyak 43% dan penyingkiran mangan sebanyak 57%. Nilai optimum untuk kepekatan pepejal terampai pula adalah 381 mg/L.

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LIST OF ABREVIATIONS

ACGIH	-	American Conference of Governmental Industrial Hygienists
COD	-	Chemical oxygen demand
CSTR	-	Continuous stirred tank reactor
EPA	-	Environmental Protection Agency
ESP	-	Electrostatic precipitator
FDA	-	Food and Drug Administration
Fe	-	Ferum
HR	-	High range
HRT	-	Hydraulic retention time
LR	-	Loading rate
Mn	-	Manganese
MSW	-	Municipal solid waste
OSHA	-	Occupational Safety and Health Administration
SBR	-	Sequencing batch reactor
SS	-	Suspended solid

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CHAPTER 1

INTRODUCTION

1 Background

Source water (ground and/or surface) may contain microbiological, chemical and radiological contaminants that must be removed or inactivated during water treatment in order to produce potable drinking water of sufficient quality (Nishijima and Speital, 2004; Kameya *et al*., 1997; Keinanen *et al*., 2004). Although ingestion of manganese (Mn) through drinking water of concentrations up to 500 µg/L has no harmful effects upon human health (WHO, 1993), its presence in drinking water at concentrations above 100 µg/L is undesirable to customers due to discoloration of the water and the subsequent staining of laundry and plumbing fixtures. The European Commission recommends an upper limit of 50 µg/L for manganese in water for drinking (Drinking Water Quality Intended for Human Consumption Belgium, 1998).

Several treatment methods to remove manganese from waste water have been reported such as trickling filter, electrodialytic process, biosorption and biofilm process. Each of these methods has some limitation in practice (Pehlivan, 2007). This research is done to prove that treating manganese by using microflora in drain is among an economically way. It also helps to determine the most applicable method in term of time and efficiency of each experiment that give more advantages and benefit. For example, modelling biological filter processes at the laboratory scale allowed the effects of defined parameter changes to be transferred to full-scale biofilters (Rowan *et al.*, 2002). By successfully modelling biofiltration and studying manganese (Mn)

bioaccumulation rates (i.e. the decrease in Mn concentration in water passing through the filter) in these filters we hoped to elucidate optimum operating parameters for the process and reduce filter maturation times from months to days. The advantages of biological treatments compared with conventional physicochemical treatments can be summarized as follows: no use of chemicals, higher filtration rates, the possibility of using direct filtration and lower operation and maintenance costs (Mouchet, 1992).

2 Objective of This Study

The main objectives of this research are:

- To study the effects of different loading rate on the manganese waste water treatment.
- To study the effects of manganese to the growth of microflora.
- To study the amount of suspended solid that will exist during the experiment.

3 Scope of Research

The scope of study in this research is to treat manganese in waste water that contains manganese was simulated with appropriate nutrients for microflora. Then, the experiments were conducted separately in reactor with loading rate. Each reactor has working volume 5 liter. The efficiency of treatment for different loading rate concentration was evaluated Mn concentration. Besides that, the effects of Mn concentration on biomass growth are defined in terms of suspended solid.

1.4 Problem Statement

Iron (Fe) and manganese cause aesthetic, organoleptic and operating problems when they are present in groundwater. These metals consume chlorine in the disinfection process and promote biofouling and microbiological induced corrosion in water networks. In groundwater, Fe and Mn are present as Fe (II) and Mn (II). The processes available for their removal are either physico-chemically or biologically based. In the absence of oxygen, iron and manganese would be reduced into soluble Fe^{2+} and Mn^{2+} states. Their presence has long been a serious problem in planning the water sources, determining the method of treatment and maintenance of the water supply system. This is evident in water treatment works with raw water source extracted from aquifers. Elevated levels of iron and manganese in drinking water will not pose health hazards apart from undesirability due to precipitation which stains clothes and utensils, corrosion of cast-iron and steel pipelines which produces "red-water" (Sawyer and McCarty, 1967). They may also impart a metallic, bitter, astringent or medical taste to water.

Manganese compounds exist naturally in the environment as solids in the soil and as small particles in water. Manganese may also be present in small dust-like particles in the air. These Mn containing particles usually settle out of the air within a few days depending on their size, weight, density, and the weather conditions. Manganese exists naturally in rivers and lakes, and is also naturally present in some underground water. Manganese can lead to the toxicity that will effect and give some problem to the crop (Bould *et* al., 1983). Algae and plankton in the water can consume some Mn and concentrate it within themselves.

In addition to occurring naturally in the environment, Mn can be introduced by human activity. Manganese can be released into the air by industry and by the burning of fossil fuels. More specifically, sources of airborne Mn include iron and steel producing plants, power plants, coke ovens, and dust from uncontrolled mining operations. Manganese released from burning a gasoline additive may also be a source of manganese in the air. Manganese from these human-made sources can enter surface water, groundwater, and sewage waters. Small Mn particles can also be picked up by

water flowing through landfills and soil. The chemical state of Mn and the type of soil determine how fast it moves through the soil and how much is retained in the soil. Maneb and mancozeb, two pesticides that contain manganese, may also add to the amount of Mn in the environment when they are applied to crops or released to the environment from packaging factories. There is information on the amount of maneb and mancozeb released into the environment from facilities that make or use these pesticides. This pesticide will induce the toxicity to the environment (Bould *et* al., 1983). However, the amount of Mn in the environment because of the release and use of these pesticides is not known.

Soluble Mn is often found in considerably greater concentrations in mine drainage waters than in unpolluted streams and groundwater (Banks *et al*., 1997). Even though there are uncertainties regarding the toxicity of manganese, recent research has shown that elevated concentrations of manganese are highly correlated to the toxicity of lake sediment pore water (Boucher and Watzin, 1999; Doyle *et al*., 2003). Besides its potential toxicity, the removal of this metal from surface and ground waters is desirable for several reasons.

In addition Mn in sources of water that are used for human consumption is undesirable because it imparts a metallic taste to water, stains laundry and water fixtures and, as Mn (IV) readily precipitates, Mn can block water distribution networks. For these reasons, the U.S. Environmental Protection Agency (EPA) has set a secondary maximum contaminant level for Mn of 0.05 mg/L. The EPA has also established guidelines limiting the concentration of Mn in acidic waters discharging from mines at maximum of 4 mg/L, as long as average discharges for a 30-day period do not exceed 2 mg/L. In the European Union, legislation under consideration may establish an Environmental Quality Standard for manganese of 0.03 mg/L.

Treatment of waste waters is essential to protect aquatic communities, and focuses on the removal of chemical and particulate contaminants. The treatment also to decrease as many as affect that can get from toxicity of manganese.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

The word Mn comes from the Latin word magnes which means magnet. In ancient time, two black mineral from magnesia were thought to differ gender which is male and female. The male magnes is attracting iron and now we know as lodestone or magnetite also gives us the term magnet. In other hand, the female magnes did not attract ore and use to decolorize glass and now known as pylorusite or manganese dioxide (Wikipedia, 2008).

In 16th century, this compound was called as magnesium by glassmaker. After that an alchemist and the glassmaker eventually had to differentiate magnesia alba (white ore) from magnesia negra (black ore). Finally the metal was isolated and called manganese. The name magnesia was then used to refer only on white magnesia alba which provide the name magnesium for that free element. In 1774, the Swedish chemist Scheele was the first person recognizes that manganese was the element and his colleague, Johan Gotties Gahn isolated the pure element by reduction of dioxide with carbon (Wikipedia, 2008).

Manganese is naturally found in many type of rock. Pure Mn is a silver-colored metal, similar to iron in its physical and chemical properties. Manganese does not exists as pure metal in environment, but is a component of more than 100 minerals, including sulfides, oxides, carbonates, silicates, phosphates, and borates. It is

combined with other elements or chemicals such as oxygen, sulfur, and chlorine to make compounds that do not evaporate (Wikipedia, 2008).

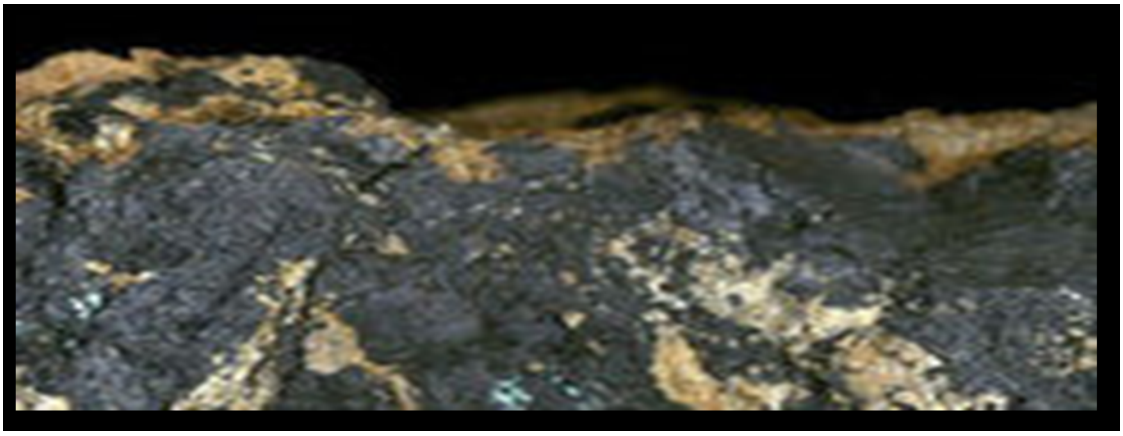

Figure 2.1: Psilomelane (manganese ore)

2.2 Metal

2.2.1 Properties of Manganese

The main ore mineral for Mn is pyrolusite (MnO_2). When Mn is alloyed with other metals like aluminum and copper, the end product is magnetic. Manganese is chemically reactive element. It easily can combine with ion in air and water. Manganese is a gray-white metal or silver, resembling iron. It is a hard metal and is very brittle, fusible with difficulty, but easily oxidized

The basic information about manganese is symbol used which is Mn and has atomic number of 25. It is transition metal in group 7, period 4 and block d. The

standard atomic weight is 54.93805 g/mole and consists of [Ar]$4s^2 3d^5$ for it electron configuration. Manganese has two different main properties which are physical properties and chemical properties which are summarize in Table 2.1, 2.2 and 2.3 (Qivx Inc.Integral Scientist Periodic Table, 2003).

Table 2.1: Physical properties of manganese

Physical properties	
Phase	solid
Density (near r.t.)	7.21 $g \cdot cm^{-3}$
Liquid density at m.p	5.95 $g \cdot cm^{-3}$
Melting point	1519 K (1246 °C)
Boiling point	2334 K (2061 °C)
Critical Temperature	4327.0 K (4054 °C)
Heat of fusion	12.91 $kJ \cdot mol^{-1}$
Heat of vaporization	221 $kJ \cdot mol^{-1}$
Specific heat capacity (25 °C)	26.32 $J \cdot mol^{-1} \cdot K^{-1}$

Table 2.2: Chemical properties of manganese

Chemical properties	
Oxidation states	+2, +3, +4, +6, +7
Electronegativity	1.55 (Pauling scale)
Atomic radius	140 pm
Covalent radius	139 pm
Vanderwaals radius	126 pm

Table 2.3: Standard reaction of manganese

Standard reactions	
oxidation	reduction
$MnO^{4-} + 4H^{+} + 3e^{-}$	$MnO_2 + 2H_2O$
$MnO^{4-} + 8H^{+} +$	$Mn^{+2} +$

5e-	$4H_2O$
$Mn^{+3} + e^-$	Mn^{+2}
$MnO^2 + 4H^+ + 2e^-$	$Mn^{+2} + 2H_2O$
$Mn^{+2} + 2e^-$	Mn

2.2.2 Environmental and Quality Issues

Toxic hazard has two types of effect which is acute (short term) exposure and chronic exposure (long term).The effects of exposure to any hazardous substance depend on the dose, the duration and how you are exposing.

Health effects from Mn are not a concern until concentrations are approximately ten times higher. The levels of Mn in groundwater from natural leaching processes can vary widely, depending upon the types of rock and minerals present at the water table. Typically, Mn concentrations from natural processes are low but can range up to 1.50 mg/L or higher. Sources of pollution, rich in organic matter (e.g., run off from landfills, composts, brush or silage piles, or chemicals such as gasoline), can add to the background level by increasing Mn release from soil or bedrock into groundwater (Division of Environmental Epidemiology and Occupational Health of Connecticut, 2001).

Manganism or Mn poisoning is a toxic condition resulting from chronic exposure to Mn and first identified in 1837 by James Couper. In initial stages of manganism, neurological symptoms consist of reduced response speed, irritability, mood changes, and compulsive behaviors. Manganism has become an active issue in workplace safety as it has been the subject of numerous product liability lawsuits

against manufacturers of arc welding supplies. In these lawsuits, welders have accused the manufacturers of failing to provide adequate warning that their products could cause welding fumes to contain dangerously high Mn concentrations that could lead welders to develop manganism (Couper, 1837).

Classical manganism presents with generally irreversible Parkinson-like symptoms (Jiang *et al*., 2006), which often progress even after exposure ends (Huang *et al*., 1993). Little is known about the potential health effects resulting from excessive ingestion exposures. Early stages of Mn toxicity include muscle tremors and can sometimes resemble Parkinson disease, which has been proposed as a sentinel among selected occupational groups. When the information got from hospital separation data for Parkinsonism between 1991 and 1995 in which New Brunswick rates were similar to other Canadian provinces (Kontakos and Stokes, 2000).

Leachates emanating from old mine workings often contain high concentrations of dissolved metals (such as Fe, Mn, Al and Zn) and can have a high acidity (Younger *et al.*, 2002; Manzano *et al.*, 1999). When these waters enter streams and rivers, Fe can rapidly become oxidized and hydrolyzed to form characteristic Fe oxyhydroxide precipitates that cover the stream bed thus suffocating much of the natural aquatic life in water courses (Jarvis and Younger, 1997). These Fe oxyhydroxides are capable of removing Mn from solution by sorption (Stumm and Morgan, 1981).

In September 2000, Mn has been found in at least 603 of the 1,517 National Priorities List (NPL) sites. It means 40% of the waste consists of manganese. As more sites are evaluated, the value will probably increase. When a substance released to the environment it will effects to biology system by breathing, eating, or drinking the substance or by skin contact.

2.2.3 Level Measurement to Discharge Manganese

Because of its potential to cause adverse health effect to the exposure, some regulations and guidelines was established for manganese. The EPA recommends that the concentration of Mn in drinking water not to be 0.005 ppm and followed by the Food & Drug Administration (FDA) for bottled water. This concentration is believed to be more than adequate to protect human health. The EPA has also established rules that set limits on the amount of Mn that factories can dump into water. EPA requires factories that use or produce Mn to report how much they dump in the environment. The Occupational Safety and Health Administration (OSHA) has set limits of 5 mg/m^3 for fume and 0.2 mg/m^3 for particulate matter as the average amounts of Mn in workplace air over 8-hour workday (OSHA, 1998). Similarly, the American Conference of Governmental Industrial Hygienists (ACGIH) has set a limit of 1 mg/m^3 for Mn fume and 0.2 mg/m^3 for the average amount of Mn, either elemental or as inorganic compounds that can be present in the air over an 8 hours workday (ACGIH, 1998).

The Food and Nutrition Board of the National Research Council (NRC) has not established a Recommended Daily Allowance for Mn because too little is known about the dietary requirements of this trace element. However, an Estimated Safe and Adequate Daily Dietary Intake (ESADDI) for Mn has been estimated as 0.3–0.6 mg/d for infants from birth to 6 months, 0.6–1 mg/d for infants aged 6 months to 1 year, 1–1.5 mg/d for children aged 1–3 years, 1–2 mg/d for children aged 4–10 years of age, and 2–5 mg/d for children aged 10 years to adult. Offices within the EPA that issue regulations for Mn are Air Quality Planning and Standards, Water Regulation and Standards, Emergency and Remedial Response, Solid Waste, and Toxic Substances.

The FDA regulates Mn concentrations in bottled water. The Department of Justice's Drug Enforcement Agency ranks potassium permanganate as an essential chemical in illegal drug production; records of sales and uses are required for amounts over 500 kg. Under Section 313 of the Emergency Planning and Community Right to

Know Act of 1986, releases of more than one pound of Mn into the air, water, and land must be reported annually and entered into the Toxic Release Inventory (TRI).

2.3 Manganese Removal Method

1 Trickling Filter

Mn(II) is readily soluble in water while Mn(III) is more unstable and has a tendency to precipitate or dissociate to Mn(II) or Mn(IV) unless chelated to another molecule. Mn(IV) is insoluble and can be detected by the presence of a visible brown or black precipitate in neutral solutions. The oxidation of Mn(II) to Mn(IV) by aeration alone is a slow process unless the pH is raised above neutrality (Cleasby, 1975; Wilson, 1980; Diem and Stumm, 1984; Hem, 1981). Therefore, Mn cannot be removed by simple aeration and precipitation. Current Mn removal methods generally required the use of strong oxidizing agents such as potassium permanganate, chlorine, hypochlorite, chlorine dioxide or ozone.

The role of microorganisms in the oxidation of Mn in drinking water production plants has attracted considerable attention (Czekalla *et al.*, 1985; Seppanen, 1988; Tamara *et al.*, 2004). A wide variety of bacteria are known to catalyze the oxidation of Mn(II). In particular, Mn oxidation can be mediated by species of several genera such as *Leptothrix, Crenotrix, Hyphomicrobium, Siderocapsa, Bacillus sp. strain SG-1, and Metallogenium* (Mouchet, 1992; Brock *et al.*, 1994; Chris *et al.*, 2001). Mn(II) oxidation rates have been shown to increase by at least 3–5 orders of magnitude in the presence of Mn(II) oxidizing bacteria (Hastings and Emerson, 1986; Tebo, 1991; Brouwers *et al.*, 1998), and several researchers have suggested that microbial oxidation is the principal pathway in the marine environment (Tebo and Emerson, 1986; Henry *et al.*, 1994). Microbial Mn(II) oxidation proceeds through indirect or direct mechanisms.

Indirect mechanisms include the production of O_2 (in photosynthesis) and of alkaline or oxidizing metabolites. Direct Mn(II) oxidation involves the microbial production of specific macromolecules (polysaccharides or proteins) catalyzing the process. The various oxidizing systems differ in many respects. For instance, the process can be catalyzed by metabolically inert spores, by cellular outer membrane components, or by bacterial sheaths. Also, according to the literature, bacterial Mn oxidation appears to be confined to outer surface coverings (Brouwers *et al.*, 2000).

Most of the work related to bacterial Mn oxidation has been focused on investigation during batch experiments (Michael and Ania, 1997; Zhang *et al.*, 2002; Boogerd and Vrind, 1987) or in natural environments (Moffett and Ho, 1996; Peter *et al.*, 1997; Alan and Tracey, 2005). Detailed information about the biological oxidation of Mn and the products produced has been also reported for the case of in situ groundwater treatment plants (Rott and Lamberth, 1993; Mettler *et al.*, 2001; Rott, Meyer and Friedle, 2002).

Several approaches have been used to express the rates of biological Mn oxidation. Studies on Mn oxidation reported in the literature use first order (Peter, 1997; Ioannis and Anastasios, 2004) or Michaelis–Menten type rate expressions (Zhang *et al.*, 2002; Moffett and Ho, 1996; Alan and Tracey, 2005) to describe the rate of Mn(II) removal, while there are only limited references concerning modeling of biological filters (Štembal *et al.*, 2005; Ioannis and Anastasios, 2004).

During Mn(II) oxidation the oxidized Mn is deposited as MnO_2 in the form of a black precipitate coating of the sand surface. It is well known that the MnO_2 layer has a catalytic effect on Mn(II) oxidation by dissolved oxygen, accelerating the process of Mn removal (Morgan, 1967). Thus, in spite of the great importance of biological processes, the chemical ones cannot be neglected.

Two pilot-scale trickling filters with fractions of various sizes of silica gravel (monolayer and multilayer filters) were constructed and tested for the biological oxidation of Mn in potable water. A series of experiments were carried out in order to study the effect of the operating parameters, namely of the Mn feed concentration and of the volumetric flow rate, as well as, the effect of the support media (monolayer and multilayer) on filter efficiency. A novel mathematical model was developed to describe the biological and autocatalytic Mn oxidation in the trickling filter. The proposed model is based on three assumptions: (a) the bacterial Mn oxidation is confined to outer surface coverings(Brouwers, 2000), (b) the bacterial cells attach on the frame of the biofilm forming a monolayer (this assumption has been also applied to the case of iron oxidation (Karamaneve, 1991; Nemati and Webb, 1999; Tekerlekopoulou, 2006) and, (c) the biofilm surface density varies at the different gravel layers due to the aggregated biomass which is deposited on the monolayer surface (this assumption was verified experimentally). Finally, mixed culture experiments were conducted in flasks to determine the net rate of biological Mn(II) oxidation and to estimate the kinetic parameters. The general working of trickling filter is represent by the Figure 2.2 as below.

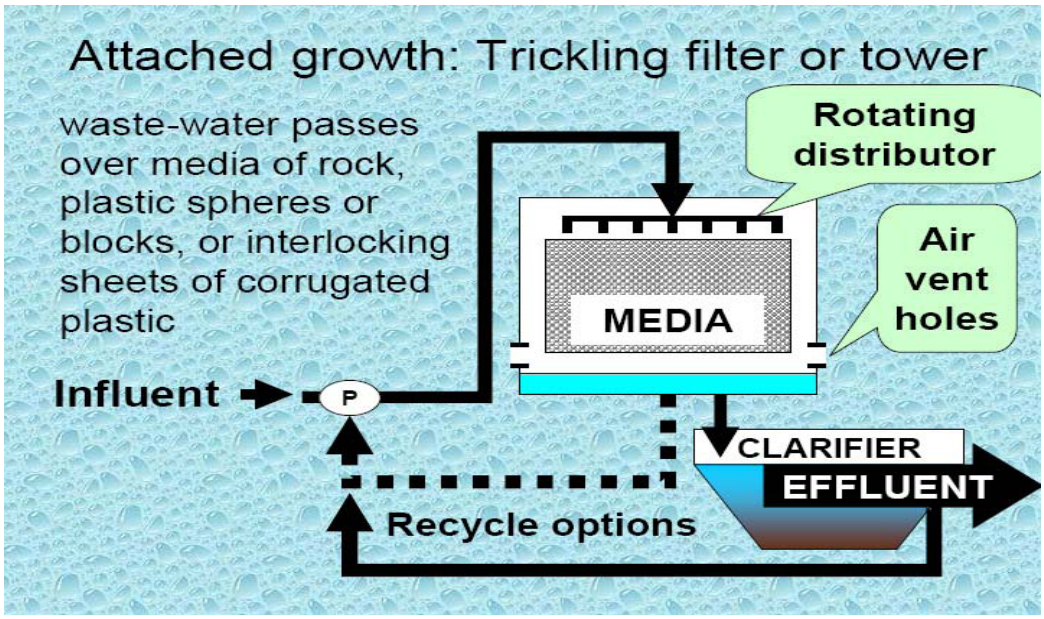

Figure 2.2: Trickling Tower

2 Electrodialytic Process

The electrodialytic remediation method was originally developed at the Technical University of Denmark (Ottosen and Hansen, 1992) for remediation of contaminated soil. The waste to be treated is saturated with water (or a solution of specific substances called "assisting agents") and placed inside the electrodialytic cell. An electric field is applied across the cell, causing the metal ions to migrate towards the electrodes, according to their charges. Selective ion exchange membranes placed between the compartments prevent the metal ions from reaching the electrodes. Instead, they are accumulated in "concentration" compartments from where they can be removed. This technique has also been applied with success to the treatment of wood waste (Ribeiro *et al.*, 2000) and biomass ash (Pedersen, 2002). Recently, some studies have been presented on the use of this technique for the treatment of electrostatic precipitator (ESP) fly ash from incineration of municipal solid waste (Pedersen, 2002).

Theoretically, the electrodialytic remediation of municipal solid waste (MSW) fly ash is possible. Fly ash particles have a porous structure and a solubility of almost 40% (w/w) (Ferreira *et al.*, 2003). A promising aspect is that the majority of the toxic metals are presumably found on the surface, as chloride compounds. This assumption is based on the fact that during combustion and gas transport, the thermodynamic conditions necessary for the formation of such compounds have occurred (Ferreira *et al.*, 2003). Since the great majority of chlorides are extremely soluble, the electrodialytic remediation may present good results.

Several conditions must be met for the successful application of this method to this material. Firstly, the solution used to prepare the initial slurry is important. The use of assisting agents during electrodialytic remediation is different for each different